

FORM PTO-1390 (Modified)
(REV 11-2000)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

TRANSMITTAL LETTER TO THE UNITED STATES

217460US0PCT

DESIGNATED/ELECTED OFFICE (DO/EO/US)

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR

CONCERNING A FILING UNDER 35 U.S.C. 371

10/030683

INTERNATIONAL APPLICATION NO.

PCT/EP00/06297

INTERNATIONAL FILING DATE

05 JULY 2000

PRIORITY DATE CLAIMED

13 JULY 1999

TITLE OF INVENTION

PRODUCTION OF FOAM SHEETS

APPLICANT(S) FOR DO/EO/US

Franz-Josef DIETZEN, et al.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (24) indicated below.
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371 (c) (2))
 - a. ☐ is attached hereto (required only if not communicated by the International Bureau).
 - b. ☒ has been communicated by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
 - a. ☒ is attached hereto.
 - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))
 - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
 - b. ☐ have been communicated by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).
10. ☐ An English language translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).
11. ☐ A copy of the International Preliminary Examination Report (PCT/IPEA/409).
12. ☒ A copy of the International Search Report (PCT/ISA/210).

Items 13 to 20 below concern document(s) or information included:

13. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
14. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
15. ☐ A **FIRST** preliminary amendment.
16. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
17. ☐ A substitute specification.
18. ☐ A change of power of attorney and/or address letter.
19. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
20. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
21. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
22. ☐ Certificate of Mailing by Express Mail
23. ☒ Other items or information:

Request for Consideration of Documents in International Search Report
Notice of Priority / PCT/IB/304

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR

10/030683

INTERNATIONAL APPLICATION NO.

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24. The following fees are submitted:

BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) :

- ☐ Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO \$1040.00
- ☒ International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO \$890.00
- ☐ International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$740.00
- ☐ International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$710.00
- ☐ International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) \$100.00

ENTER APPROPRIATE BASIC FEE AMOUNT =

\$890.00

Surcharge of \$130.00 for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492 (e)).

\$0.00

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE
Total claims	1 - 20 =	0	x \$18.00
Independent claims	1 - 3 =	0	x \$84.00

\$0.00

\$0.00

Multiple Dependent Claims (check if applicable). ☐

\$0.00

TOTAL OF ABOVE CALCULATIONS =

\$890.00

☐ Applicant claims small entity status. See 37 CFR 1.27). The fees indicated above are reduced by 1/2.

\$0.00

SUBTOTAL =

\$890.00

Processing fee of \$130.00 for furnishing the English translation later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492 (f)).

\$0.00

TOTAL NATIONAL FEE =

\$890.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable). ☐

\$0.00

TOTAL FEES ENCLOSED =

\$890.00

Amount to be:
refunded \$
charged \$

- a. ☒ A check in the amount of \$890.00 to cover the above fees is enclosed.
- b. ☐ Please charge my Deposit Account No. _____ in the amount of _____ to cover the above fees. A duplicate copy of this sheet is enclosed.
- c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 15-0030 A duplicate copy of this sheet is enclosed.
- d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. **Credit card information should not be included on this form.** Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:



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Surinder Sachar
Registration No. 34,423

SIGNATURE

Norman F. Oblon

NAME

24, 618

REGISTRATION NUMBER

DATE

Jan 14 2002

Production of foam sheets

The invention relates to a process for producing foam sheets with
5 a density of from 20 to 200 g·l⁻¹ and with a cross section of at least 50 cm² based on styrene polymers which comprise graphite particles to reduce thermal conductivity.

Extruded polystyrene foams (XPS) are widely used to insulate
10 buildings and parts of buildings. For this application the foam sheets have to have very low thermal conductivity. Nowadays, halogen-free blowing agents, preferably CO₂-containing blowing agent mixtures, are used for producing XPS sheets in order to protect the environment. However, CO₂ diffuses out of the foam
15 cells significantly more rapidly than fluorine-containing gases and is replaced by air. The thermal conductivity of XPS sheets produced with CO₂-containing blowing agents is therefore somewhat higher than that of XPS sheets produced with fluorohydrocarbons. It is known from EP-A 863 175 that adding graphite particles can
20 reduce thermal conductivity during XPS production. However, it has been found that the nucleating action of the graphite here gives a very fine-celled foam, meaning that it is not possible to produce thick sheets. It is an object of the present invention, therefore, to provide very thick XPS sheets with low thermal
25 conductivity.

We have found that this object is achieved if, during the production of foam sheets with a density of from 20 to 200 g·l⁻¹ and with a cross section of at least 50 cm³ by extrusion and
30 foaming of a mixture made from a styrene polymer, from 3 to 15% by weight of a volatile blowing agent and from 0.2 to 10% by weight of graphite particles, based in each case on the styrene polymer, if desired with conventional additives, the blowing agent used comprises a mixture made of
35 from 95 to 30% by weight of CO₂,
from 5 to 70% by weight of H₂O, and
from 0 to 60% by weight of a volatile organic compound.

WO 93/25 608 describes the production of XPS foams with a bimodal
40 foam structure, using a blowing agent mixture made from CO₂, H₂O and C₂H₅OH, with addition of carbon black during extrusion. The water content in the blowing agent mixture is said to be responsible for the bimodal foam structure, and the carbon black additive is intended to reduce thermal conductivity. However, it
45 has been found that a bimodal foam structure is disadvantageous since it makes operations on the foam sheets, e.g. sawing, milling, cutting or embossing, more difficult. Surprisingly, it

has been found that adding graphite instead of carbon black not only further lowers the thermal conductivity but also can prevent the formation of a bimodal foam structure. WO 93/25 608 also gives no indication that the water content in the blowing agent mixture permits thick sheets to be produced.

WO 94/09 975 teaches that XPS foams with a monomodal foam structure are obtained when a CO₂/H₂O blowing agent mixture is used if the water-solubility of the polymer melt is increased. No mention is made in this publication of adding graphite particles in the production of XPS.

For the purposes of this invention, styrene polymers are polystyrene and copolymers of styrene in which there is at least 50% by weight of copolymerized styrene. Examples of possible comonomers are α -methylstyrene, ring-halogenated styrenes, ring-alkylated styrenes, acrylonitrile, (meth)acrylates of alcohols having from 1 to 8 carbon atoms, N-vinyl compounds, such as vinylcarbazole, maleic anhydride or else small amounts of compounds which contain two polymerizable double bonds, such as butadiene, divinylbenzene or butanediol diacrylate.

The foam sheets comprise from 0.2 to 10% by weight of graphite particles uniformly distributed, preferably from 1 to 8% by weight of graphite with a particle size of from 1 to 100 μ m, preferably from 2 to 20 μ m.

It is advantageous to use flame retardants in producing the XPS, preferably from 0.5 to 5% by weight of organic bromine compounds with a bromine content of more than 70%, e.g. hexabromocyclododecane, preferably together with from 0.1 to 0.5% by weight of an organic compound which has labile C-C bonding or labile O-O bonding, for example dicumyl peroxide or preferably dicumyl.

Other conventional additives and/or auxiliaries which may be added to the polystyrene matrix are antistats, stabilizers, dyes, fillers and/or nucleating agents, in the usual amounts.

The blowing agents used comprise from 3 to 15% by weight, preferably from 4 to 12% by weight, based on the styrene polymer, of a mixture made of from 95 to 30% by weight, preferably from 90 to 40% by weight, of CO₂, from 5 to 70% by weight, preferably from 10 to 60% by weight, of H₂O, and

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from 0 to 60% by weight, preferably from 0 to 30% by weight, of a volatile organic compound.

The volatile organic compound preferably has a boiling point of
5 from 0 to 100°C, in particular from 30 to 80°C. Examples of suitable compounds are alcohols, aliphatic hydrocarbons, ketones and ethers. Ethanol is particularly preferred.

An advantage of adding water to the blowing agent mixture is that
10 the amount of combustible organic blowing agents can be reduced or even dispensed with entirely. When graphite is used instead of carbon black as an agent for reducing the thermal conductivity of the foam sheets, then even when H₂O is used as blowing agent a monomodal foam structure is obtained. It appears that the
15 concomitant use of water as blowing agent reduces the nucleating effect of the graphite, so that it is also possible to produce thick foam sheets.

The percentages mentioned in the examples are based on weight.

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Examples

The foam samples are extruded on a tandem plant. This is composed of a twin-screw ZSK53 extruder and a single-screw cooling
25 extruder (KE 90). Polymer and additives were introduced to the twin-screw extruder. The polymer was melted at 210°C and the mixture of the blowing agents was injected in its entirety at a single point. The melt comprising blowing agents was then cooled in the second extruder to 120-135°C, the temperature needed for
30 foaming. The throughput was 50 kg/h, and the die had a width of 70 mm. The height of the die gap was 3 mm. Graphite powder (AF spez. 96-97, average particle size 6 µm, from Graphitwerk Kropfmühle) was added to the polystyrene. The makeup of the blowing agent and the results are shown in the table.

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Table

Example	Blowing agent mixture %			Additives %			Density g.l ⁻¹	Thickness mm	Th. cond. λ mW·m ⁻¹ ·K ⁻¹
	CO ₂	H ₂ O	C ₂ H ₅ OH	Graphite	Carbon black	Talc			
1	2	3	-	-	-	2	42	66	38
2	2	3	-	-	2	-	43	67	35
3	2	3	-	2	-	-	46	72	32
4	2	2	1	2	-	-	44	70	32
5	2	-	3	2	-	-	56	48	33

Examples 3 and 4 are according to the invention

The foam in Example 2 has a bimodal foam structure.

We claim:

A process for producing foam sheets with a density of from 20 to
5 200 g·l⁻¹ and with a cross section of at least 50 cm³ by extrusion
and foaming of a mixture made from a styrene polymer, from 3 to
15% by weight of a volatile blowing agent and from 0.2 to 10% by
weight of graphite particles, based in each case on the styrene
polymer, if desired with conventional additives, which comprises
10 using a volatile blowing agent which is a mixture made of
from 95 to 30% by weight of CO₂,
from 5 to 70% by weight of H₂O, and
from 0 to 60% by weight of a volatile organic compound.

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Declaration, Power of Attorney

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0050/050465

We (I), the undersigned inventor(s), hereby declare(s) that:

My residence, post office address and citizenship are as stated below next to my name,

We (I) believe that we are (I am) the original, first, and joint (sole) inventor(s) of the subject matter which is claimed and for which a patent is sought on the invention entitled

PRODUCTION OF FOAM SHEETS

the specification of which

☐ is attached hereto.

☐ was filed on _____ as

Application Serial No. _____

and amended on _____.

☒ was filed as PCT international application

Number _____
PCT/EP00/06297

on _____
July 5, 2000

and was amended under PCT Article 19

on _____ (if applicable).

We (I) hereby state that we (I) have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

We (I) acknowledge the duty to disclose information known to be material to the patentability of this application as defined in Section 1.56 of Title 37 Code of Federal Regulations.

We (I) hereby claim foreign priority benefits under 35 U.S.C. § 119(a)–(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed. Prior Foreign Application(s)

Application No.	Country	Day/Month/Year	Priority Claimed
19932619.3	Germany	13 July 1999	<input checked="" type="checkbox"/> Yes <input type="checkbox"/> No

We (I) hereby claim the benefit under Title 35, United States Codes, § 119(e) of any United States provisional application(s) listed below.

(Application Number)

(Filing Date)

(Application Number)

(Filing Date)

We (I) hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

Application Serial No.

Filing Date

Status (pending, patented,
abandoned)

_____	_____	_____
_____	_____	_____
_____	_____	_____
_____	_____	_____

And we (I) hereby appoint:

Norman F. Oblon, Registration Number 24, 618;
Marvin J. Spivak, Registration Number 24, 913;
Gregory J. Maier, Registration Number 25, 599;
William E. Beaumont, Registration Number 30, 996;
Steven B. Kelber, Registration Number 30, 073;
Jean-Paul Lavallee, Registration Number 31, 451;
Timothy R. Schwartz, Registration Number 32, 171;
Stephen G. Baxter, Registration Number 32, 884;
Richard L. Treanor, Registration Number 36, 379;
Robert W. Hahl, Registration Number 33, 893, our (my) attorneys, with full

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powers of substitution and revocation, to prosecute this application and to transact all business in the Patent Office connected therewith; and we (I) hereby request that all correspondence regarding this application be sent to the firm of **OBLON, SPIVAK, McCLELLAND, MAIER & NEUSTADT, P. C.**, whose Post Office Address is: Fourth Floor, 1755 Jefferson Davis Highway, Arlington, Virginia 22202.

We (I) declare that all statements made herein of our (my) own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

1-0
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Declaration

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5-0

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